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Positron Annihilation Spectroscopy on LiBH_4 and $\text{LiBH}_4\text{:LiI}$ superionic lithium conductors

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Rechargeable lithium-ion batteries are standard in today's consumer portable electronics but substantial improvements are needed to achieve higher energy densities, safer use, longer lifetimes and reduce prices to meet the demand for example in the transport sector and storage of electrical power. Conventional Li-ion batteries use electrolytes made of organic liquids or gels. They have high Li^+ conductivity, but are flammable, causing safety issues, and allow lithium dendrite formation at the electrode-electrolyte interface. These dendrites cause a decrease of the cell capacity, reducing their lifetime, not to mention the possibility of hazardous short circuits.

Lithium Borohydride, LiBH_4 , is potentially interesting as a solid state electrolyte for Li-ion batteries. It consists of a lattice of Li^+ cations and BH_4^- anions and displays high lithium mobility, not at room temperature but above ~ 390 K where a transition to a high temperature hexagonal structure occurs. This hexagonal, highly Li^+ conducting phase, can be stabilized at room temperature by lithium halides in solid solutions, in particular lithium iodide.

Considering that the Li^+ conductivity is strongly influenced by the presence of Li-ion vacancies we have carried out a series of positron lifetime measurements in order to take advantage of the defect sensitivity of positron annihilation spectroscopy.

In coarse-grained, as-prepared LiBH_4 the o-Ps lifetime shows a sharp, reversible increase at the phase transition. In ball-milled, fine-grain material the lifetime is longer than for the as-prepared material at all increasing temperatures from 296 K to 420 K and shows no phase transition, while a subsequent decrease of the temperature results in a reduction of the lifetime and an indication of a transition, probably due to the heat treatment at the highest temperatures. In LiBH_4 stabilized with LiI a gradual, reversible lifetime increase is observed in the whole temperature range.

Particularly interesting is the finding that the variations of the positron lifetime and the Li^+ conductivity are qualitatively identical.

Details of the measurements and their interpretation will be presented at the conference.